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Evaluation of the Strategic Value of Fully Burnt PBMR Spent Fuel

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Executive Summary

The spent fuel from the PBMR-400 reactor has been evaluated for suitability for further utilization in nuclear applications. The approach taken in this evaluation is detailed and justified and the conclusions of the study are presented.

The PBMR-400 spent fuel is considered against criteria derived from the INFCIRC/66, INFCIRC/153 (Corrected) and the IAEA Safeguards Manual. The principal conclusion of this study is that per the criteria of INFCIRC/66, INFCIRC/153 (Corrected) and the IAEA Safeguards Manual, the fully burnt spent fuel from the operation of the South African PBMR-400 pebble bed reactor must remain under safeguards until such time as it is (chemically) treated to separate the fissile materials from it. **The spent fuel does not meet the criteria for termination of safeguards on measured discards**. However, the PBMR spent fuel will not require safeguards more stringent than those that apply to spent fuel from the current generation of light water reactors or to the spent fuel of other high temperature gas-cooled reactors currently being considered for development by some entities worldwide.

For all cases the Pu-238 content is very far from the 80% content required for termination of safeguards on measured discards. The fissile isotopes contents per pebble (especially Pu-239) are small but still sufficiently large to be attractive to a would-be proliferant. The fissile isotopes contents of the spent fuel, predicted through modeling, are roughly of the same magnitude as the fissile content of fresh light water reactor fuel. As a consequence, with a small amount of processing the PBMR-400 spent fuel could be used as a source of fuel for another reactor, albeit not another PBMR. For proper perspective, it must be noted that the residual fissile isotopes content of prismatic block HTR concepts is slightly higher than that of spent PBMR-400 fuel, implying at least similar safeguard constraints.

The reprocessing of TRISO-based spent fuel is a difficult and expensive undertaking. Notwithstanding the expense, the fissile content of TRISO-based spent fuel cannot be viewed as irrevocably irretrievable from the point of view of safeguards.

The conclusion of this study is that the fully burnt spent fuel from the PBMR-400 reactor does not meet the IAEA criteria for termination of safeguards on measured discards. Therefore safeguards should be maintained on such spent fuel until it has been treated to meet said IAEA criteria. For completeness, it is important to note that spent fuel from prismatic block HTRs would have to be subjected to at least the same restrictions.

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1. Introduction

The IAEA needs to determine the value of imposing safeguards on the spent fuel storage at the Pebble Bed Modular Reactor (PBMR) planned for construction in the Republic of South Africa.

The PBMR will use hundreds of thousands of fuel elements in the shape of small spheres (6 cm in diameter). The PBMR plant design calls for the storage on site of all the spent fuel generated during the whole life of the reactor, expected to span 40 years [i]. The spent fuel storage system is designed (or to be designed) for a functional life of 80 years [i].

If it is determined that the spent fuel contains materials of interest to a would-be proliferant, then safeguards would have to be imposed and maintained until the spent fuel elements are processed into a form and composition that no longer requires safeguards. The problem addressed in this report is the determination of the strategic value of the spent fuel to such a would-be proliferant. The report is meant to answer the question of

> "Whether the fully-burnt PBMR fuel would satisfy, or come close to satisfying, currently applied criteria for termination of safeguards on measured discards."

Because it is anticipated that PBMR fuel will be burnt to a much higher level than current generation of light water reactor fuel, and because it is generally believed that an effective technology for the reprocessing of PBMR fuel has not yet been developed, this is a legitimate question.

The question to be answered requires a two-fold assessment:

- Assess the contents of the spent fuel from the perspective of usefulness to a would-be proliferant, i.e. determine if the contents of spent fuel possess isotopic characteristics that make it suitable for use, either immediate or future, in a nuclear weapons-procurement effort (including if additional refinements are required).

- If determined to be suitable for nuclear weapons-applications or for feed into a nuclear weapons-procurement effort, then further determine whether the materials contained within the spent fuel elements can be practically extracted/recovered.

These two assessment efforts were carried out in parallel and the outcome is presented in this report.

In the next section (Section 2), the criteria applicable to the determination of the strategic value of spent PBMR fuel are identified. The following section presents the assessment of PBMR spent fuel against the first set of criteria, namely the isotopics of fully burnt spent fuel. Section 4 presents a brief assessment of the state of the art of reprocessing TRISO-type fuel and hence of the ability to recover from it the materials of interest. Section 5 identifies the limits of the current study, including an outlining of the scenarios that have not been considered. It also includes a brief discussion of the limitations in the state-of-the-art methods used in the study. Section 6 presents the main recommendations that stem from the results of the study and concludes this report. Section 7 lists the references cited in the text of the report.

The principal conclusion of this study is that per the criteria of INFCIRC/66, INFCIRC/153 (Corrected) and the IAEA Safeguards Manual, the fully burnt spent fuel from the operation of the South African PBMR-400 pebble bed reactor must remain under safeguards until such time as it is (chemically) treated to separate the fissile materials from it. **The spent fuel does not meet the criteria for termination of safeguards on measured discards**. However, the PBMR spent fuel will not require safeguards more stringent than those that apply to spent fuel from the current generation of light water reactors or to the spent fuel of other high temperature gas-cooled reactors currently being considered for development by some entities worldwide.

2. Applicable Criteria and their Consequences

The criteria applicable to the determination of the acceptability of termination of safeguards for the PBMR spent fuel are identified in this section. The first item addressed is whether the PBMR spent fuel can be considered a "measured discard." The remaining criteria that are considered for applicability stem either from IAEA documentation or from explicit instructions received from ISPO during the awarding process for this project. All are reviewed in this section.

2.1 Measured Discards

Per the IAEA Safeguards Manual, referencing INFCIRC/153 (Corrected), measured discards are defined as "nuclear material, which has been measured, or estimated on the basis of the measurements, and disposed of in such a way that it is not suitable for further nuclear use."

The above definition for measured discards is somewhat ambiguous when its application to spent nuclear fuel is contemplated. Indeed, spent fuel is usually in a form that is not suitable for *immediate*, *in its as-is-state*, further nuclear use, but future use cannot be precluded for spent fuel. The future nuclear use may of course require additional intermediate or preliminary steps. If the definition is meant to imply unsuitability for *all* future nuclear use, then it is clear that the definition is not proper for the PBMR spent fuel, as is made clear in Section 4 of this report since it can be shown there that the spent fuel is not a form in which nuclear useable materials are irrevocably irretrievable. Furthermore, the criteria for allowing termination of safeguards given in the Safeguards Manual (SMR 2.14), Section 5.2, clearly apply to wastes in the commonly accepted sense that excludes spent fuel. In particular, the concentrations of uranium and plutonium given in Tables 1 and 2 of that section (SMR 2.14) of the manual are clearly meant for wastes from reprocessing of spent fuel but not for the spent fuel proper or for the fissile materials recovered through reprocessing.

In light of the above conclusion that the PBMR spent fuel does not strictly meet the definition for a "measured discard," the question remains as to whether safeguards must be maintained in accordance with other relevant rules or criteria or whether the safeguards could be discontinued. Those criteria are discussed below.

2.2 Criteria in the IAEA Safeguards Manual and INFCIR/66 and INFCIRC/153 (Corrected)

The discussion on Section 2.1 above clearly demonstrate that the criteria contained in the portion of the IAEA Safeguards Manual that was supplied to the INL by ISPO does not apply to the spent fuel from the PBMR-400 reactor.

SMR 2.14 (i.e. Policy Paper 2.14 of the IAEA Safeguards Manual) summarizes the criteria for termination of safeguards on measured discards as given in INFCIR/66 and INFCIRC/153

(Corrected). Although it is determined above that the PBMR spent fuel does not meet the definition of a measured discard, we subject it in this study to an evaluation against the existing criteria. These criteria are spelled out in two statements regarding termination of safeguards and corresponding to INFCIRC/153 (Corr.), para.11 and to INFCIRC/66/Rev. 2, para.26c, respectively. These two statements are:

i) The agreement should provide that safeguards shall terminate on nuclear material subject to safeguards there under upon determination by the Agency that it has been consumed, or has been diluted in such a way that it is no longer usable for any nuclear activity relevant from the point of view of safeguards, or has become practicably irrecoverable (see INFCIRC/153 (Corr.), para. 11),

ii) Nuclear material shall no longer be subject to safeguards after the Agency has determined that it has been consumed, or has been diluted in such a way that it is no longer usable for any nuclear activity relevant from the point of view of safeguards, or has become practicably irrecoverable (see INFCIRC/66/Rev. 2, para.26c).

In these two statements the three primary criteria are that the material was “consumed” or “diluted” or that it has become “practicably irrecoverable.” The threshold for these changes to make termination of safeguards allowable is that due to its final physical properties the material is no longer useful for any activity relevant from the point of view of safeguards.

2.3 Recommendation and Course of Action

The spent fuel from the PBMR-400 reactor must be characterized with respect to the criteria identified in Section 2.2, regardless of its categorization as a measured discard (or its non-categorization as such a material). The first criterion, namely having been “consumed,” implies the need to determine the final fissile materials content of the PBMR-400 spent fuel, then compare that content to a valid threshold (to be identified in the results section) that defines future usefulness. The threshold should incorporate the total amount of fissile materials present as well as their isotopic distribution. The same calculations should allow the generation of data that allow the determination of the level of relevant materials dilution in the spent fuel.

If the materials were determined to be consumed or diluted sufficiently to meet the threshold conditions (that will be spelled out in the results section), then the determination of the irretrievability would not be necessary. However, if, instead, neither of the two first criteria is met, whether the materials are in a physical and/or chemical form that allows retrieval will determine the necessity (or absence of necessity) for continued safeguards.

In light of the criteria that are identified above, it is clear that the contents of the PBMR-400 spent fuel must be characterized with respect to their fissile materials contents and with respect to the ease or difficulty with which said materials could be recovered for further or future nuclear uses. The course of action chosen in this project is the one that addresses these characterizations using quantitative predictions in one case, and using a limited survey of the relevant literature enriched by INL experts’ opinions in the other. The course of action is further detailed in Section 2.5.

2.4 Instructions in Letter from ISPO/IAEA

The letter from ISPO/IAEA [ii] that requested the proposal for this project was explicit in requesting a determination of the residual strategic value of the PBMR-400 spent fuel. The letter also requested a determination of the validity of a possible decision to terminate safeguards. Both of these requests implicitly assumed full compliance with all applicable IAEA safeguards criteria and explicitly requested a determination against the criteria applicable to measured discards. The latter criteria were discussed in Section 2.1 and found not to apply. The former have been discussed in Section 2.2 and their implications identified. By addressing the criteria from the ISPO/IAEA request, this report constitutes a deliverable that is fully responsive to the instructions received from ISPO.

2.5 Consequences: translation of criteria into a course of action

The course of action chosen in this work is the one that was anticipated in the proposal that was submitted and that resulted in the award. For completeness and in order to avoid oversights, all the criteria that could have applied to the determination of the strategic value of the spent fuel from the PBMR-400 reactor have been reviewed. Since the outcome of this review did not differ from the anticipated course of action outlined in the proposal, further concurrence from the ISPO/IAEA sponsors was not sought and the actions leading to the determination were carried out.

The criteria for the determination of the need for continued safeguards on PBMR-400 spent fuel identified above are translated into two specific tasks in this project. In the first task the isotopics and the concentration of fissile materials in the PBMR spent fuel must be examined in order to determine whether the spent fuel in its as-is-state could be used in any nuclear process or nuclear activity relevant from the point of view of safeguards. The second task is a determination of the ease or difficulty of retrieval of such materials from the spent fuel.

2.5.1 Determination of Isotopics and Concentration of Fissile Materials

It is generally recognized that the isotopic distribution of plutonium intended for weapons production must be very strictly controlled. Some compositions can result in significant technological difficulties in the manufacturing of a weapon, with some mixes making a ***practical*** weapon simply impossible.

The criterion that will be applied in this work derives directly from IAEA texts and pertains to the ^{238}Pu isotopic fractional content of the plutonium within the spent fuel. Per IAEA criteria, if the ^{238}Pu content is larger than 80% of all the plutonium inventory of the spent fuel, then safeguards could be terminated, as the material is deemed unsuitable to the manufacturing of nuclear explosives in effective configurations.

The strict adherence to the 80% ^{238}Pu content requirement for termination of safeguards has been criticized by many authors. Proponents of a lower limit have put forward defensible arguments in favor of such a lesser limit as well as arguments in favor of a criterion that takes into account the mix of all plutonium isotopes in formulating a condition for termination of safeguards. Despite those advocates of a different limit, the present works adheres strictly to the criterion of a ^{238}Pu content for termination of safeguards. This choice is made in order to be most responsive to the needs of the IAEA and because there could be significant physics reasons for even this criterion to be insufficiently conservative, although the details underlying this remark are beyond the scope of this project.

The first task is to determine the isotopics of the plutonium contained in the spent fuel from the PBMR-400 reactor and to compare that content to the 80% ^{238}Pu criterion.

In addition to determining the isotopic distribution of the plutonium in the PBMR spent fuel, the actual average content of plutonium per pebble will also be determined. This is important because too small a content, or conversely too high a dilution, would imply a lower value to a would-be proliferant.

The plutonium contents per unit mass of spent fuel and the plutonium isotopic distribution will be compared to the criteria for termination of safeguards on measured discards and the results of the comparison will be reported.

2.5.2 Determination of Ease/Difficulty of Retrieval

The plutonium contained in the spent fuel pebbles is useless to a would-be proliferant if it cannot be extracted and retrieved for further use. It is therefore important to determine whether ***practicable*** methods can be deployed for the retrieval of *sufficiently large* quantities of plutonium from PBMR spent fuel. Until very recently, the relevant literature seemed to point toward important difficulties in trying to reprocess PBMR spent fuel. In the very recent past, new information has appeared that contradicts this traditional claim of "impossibility to reprocess TRISO fuel." Furthermore, other information may also contradict this heretofore widely held belief.

This task will consist in a brief survey of the literature enriched by the input of subject matter expert opinion from INL. The report will include the findings from the literature survey with incorporation of the experts' opinions.

3. Isotopics of PBMR Spent Fuel Pebbles

In order to produce an assessment of the isotopics of PBMR spent fuel, a simple operational model of the PBMR-400 reactor will be constructed using the ***current version*** of the PEBBED code, albeit with some modification to track all the relevant isotopes. An "average" pebble that is fully depleted will be used as the model. The plutonium content of that pebble and the isotopic distribution of that plutonium will be reported and compared to those of weapons grade plutonium, and of light water reactors spent fuel as well. From these comparisons, the level of attractiveness, to a would-be proliferant, of the plutonium within the spent fuel would be assessed and reported. In addition to the plutonium content, the uranium content is reported. The latter includes a breakdown between the U^{235} and U^{238} isotopes in order to characterize the residual enrichment.

The ***current version*** of the PEBBED code is capable of a high level of fidelity in the modeling of the PBMR-400 reactor. However, while it produces the best possible results about the asymptotic burnup and loading pattern of the PBMR reactor, it does not allow the analysis of intermediate stages, prior to the establishment of the asymptotic (or equilibrium) fueling pattern. This should not be a major concern since the asymptotic pattern will prevail during most of the lifetime of the facility. Furthermore, the fully burnt spent fuel will have experienced burnup to the maximum allowable, regardless of the state of the reactor core away from the asymptotic pattern and the spectrum that would have been experienced by the spent fuel would not be significantly different from that of the asymptotic core. The information that will be produced ***with the current version*** will be ***fully sufficient*** for answering with adequate confidence the questions identified in Section 1 above. In contrast, if sophisticated proliferation scenarios were to be considered and if their signatures were to be identified, the code would have to be augmented in a separate few person-months effort. Such a development of a full fidelity model is well within the capability of the INL, but would be incompatible with an effort that adds up to a total investment of the order of one person-month. Also, if more details were required about fully burnt fuel discharged during the approach to asymptotic loading pattern phase, a new capability will be required. That capability is currently under development at the INL.

3.1 Methodology

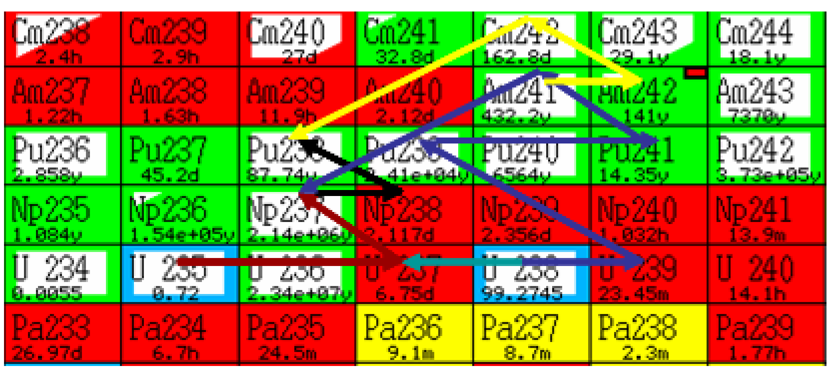
3.1.1 The PEBBED code and its capabilities

The PEBBED code [iii], which defines the state of the art in pebble reactor design and analysis methods, has been under development at the INL for longer than the past six years. The code is capable of very sophisticated operation scenarios modeling and of optimization of the fuel cycle and of the operation mode of the reactor. The code has been used, for example, to demonstrate that a pebble bed reactor of power greater than 600 MWth could be designed that is also passively safe [iv]. Until the PEBBED code became available, it was commonly believed that a passively safe pebble reactor design could not exceed the "natural" limit of 400 MWth. Currently the PEBBED code is being used in the design of the Next Generation Nuclear Plant (NGNP), a Very High Temperature Reactor (VHTR) [v]. Concurrently, the code is undergoing additional improvements for greater efficiency and fidelity. In the recent past, the PEBBED code was used to study the nonproliferation characteristics [vi,vii] of the PBMR-268 reactor design (the predecessor of the PBMR-400 design now under development in the Republic of South Africa).

3.1.2 Chains modeled in PEBBED for this study

Prior to this project, the PEBBED code was not required to generate a precise accounting for the generation and destruction of the 238 isotope of plutonium (Pu-238). For this study, the capability to compute the inventory of Pu-238 in spent fuel (along with other isotopes) was added to the code. The chains that lead to the formation (or destruction) of Pu-238, as now modeled in the PEBBED code are shown and discussed below. Isotopes that have extremely short half-lives are bypassed.

Production of Pu-238 largely comes from the capture of a neutron by Np-237 and subsequent beta decay of Np-238. The Np-237 isotope has a thermal capture cross section (σ_c) of 170 b and a resonance integral (I_c) of 730 b. This is shown as the black path on the chart, below.



The relevant reactions for these paths are:

$$Np_{93}^{237} + n_0^1 \rightarrow Np_{93}^{238} \qquad \sigma_c = 170\text{b}, \mathrm{I_c} = 730\text{b}$$
$$Np_{93}^{238} \rightarrow Pu_{94}^{238} + \beta_{-1}^{0} \qquad t_{1/2} = 2.117d$$

Since production of Pu-238 is primarily from this path that starts with Np-237, then all paths that lead to this isotope should be accounted for. These are enumerated below.

(1) The first such path is through successive neutron captures starting from U-235, followed by decay to Np-237. This is shown as the red path on the chart. This path dominates with a ratio of ~6:1 in

light water reactor spectra, and therefore is expected to be similarly significant in the PBMR-400 thermal spectrum (though the fuel kernels are exposed to a harder spectrum than LWR fuel). This path was already modeled in PEBBED prior to this project.

There are three relevant principal transmutation reactions that occur in LEU in a flux field:

$$U_{92}^{235} + n_0^1 \rightarrow U_{92}^{236} \qquad \sigma_c = 99\mathrm{b}, \mathrm{I}_c = 140\mathrm{b}$$
$$U_{92}^{236} + n_0^1 \rightarrow U_{92}^{237} \qquad \sigma_c = 5.1\mathrm{b}, \mathrm{I}_c = 360\mathrm{b}$$
$$U_{92}^{237} \rightarrow Np_{93}^{237} + \beta_{-1}^0 \qquad t_{1/2} = 6.75d$$

(2) A second path feeding the production of Np-237 starts with U-238. This isotope undergoes an (n, 2n) reaction. This is shown with the green arrow on the chart. This path is probably significant in fast reactors, but in that case most of the nuclides formed would still not remain in the spent fuel, as they would be burnt. This path is not modeled within PEBBED. The transmutation chain for this path is:

$$U_{92}^{238} + n_0^1 \rightarrow U_{92}^{237} + 2n_0^1 \qquad \sigma_{\mathrm{n,2n}} = 0.00095 \text{ b above } 0.1\mathrm{MeV}$$
$$U_{92}^{237} \rightarrow Np_{93}^{237} + \beta_{-1}^0 \qquad t_{1/2} = 6.75d$$

(3) The third path is through Am-241 alpha decay (blue arrow on the chart): this is a comparatively small contributor but still a non-negligible one for high-burnup fuel because of the Pu buildup. Note: Am-241 continues to be produced within spent fuel outside of the core. The relevant transmutation equations for this production path are:

$$U_{92}^{238} + n_0^1 \rightarrow U_{92}^{239} \qquad \sigma_c = 2.68\mathrm{b}, \mathrm{I}_c = 277\mathrm{b}$$
$$U_{92}^{239} \rightarrow Np_{93}^{239} + \beta_{-1}^0 \qquad t_{1/2} = 23.5m$$
$$Np_{93}^{239} \rightarrow Pu_{94}^{239} + \beta_{-1}^0 \qquad t_{1/2} = 2.355d$$
$$Pu_{94}^{239} + n_0^1 \rightarrow Pu_{94}^{240} \qquad \sigma_c = 271\mathrm{b}, \mathrm{I}_c = 200\mathrm{b}$$
$$Pu_{94}^{240} + n_0^1 \rightarrow Pu_{94}^{241} \qquad \sigma_c = 290\mathrm{b}, \mathrm{I}_c = 810\mathrm{b}$$
$$Pu_{94}^{241} \rightarrow Am_{95}^{241} + \beta_{-1}^0 \qquad t_{1/2} = 14.4y$$
$$Am_{95}^{241} + \alpha_2^4 \rightarrow Np_{93}^{237} \qquad t_{1/2} = 432.7y$$

There are a number of minor branches of these chains that may yield miniscule quantities of Pu-238. One such branch of the U-238 transmutation chain, modeled in PEBBED, involves the alpha decay of Cm-242 as follows:

$$Am_{95}^{241} + n_0^1 \rightarrow Am_{95}^{242} \qquad \sigma_c = 50\mathrm{b}, \mathrm{I}_c = 550\mathrm{b}$$
$$Am_{95}^{242} \rightarrow Cm_{96}^{242} + \beta_{-1}^0 \qquad t_{1/2} = 16.0y$$
$$Cm_{96}^{242} + \alpha_2^4 \rightarrow Pu_{94}^{238} \qquad t_{1/2} = 163d$$

3.2 Results

Using the suite of PEBBED and its support codes, and including the changes necessary to model all of the important relevant nuclides, the discharge isotopics of PBMR-400 spent fuel have been computed for two modes of operation. The first one assumes the reactor is operating with all control rods out and the second assumes control rods are partially inserted at the top portion of the core. The former operation mode is plausible because the pebble bed reactor concept requires very little (and theoretically no) excess reactivity, as fuel is added at a nearly constant rate. If no interim shutdown of the reactor is expected, and the reactor is used as base load generator and not in a load-follow mode, then the hypothesis of fully withdrawn control rods is actually realistic. The latter mode, with some control rods partly inserted into the core, is justified when the reactor is required to be able to resume operation promptly after a short shutdown or to operate in a load-follow mode. The excess reactivity held down by the partly inserted control rods is necessary to overcome the effect of Xe buildup. The partial insertion of the control rods is also desirable if it is intended to flatten the axial power shape and to shift the peak of the power generation profile lower into the core. Results for these two operation modes are presented in Table 1 and then discussed in turn. A detailed model of the PBMR-400 has been built for the coupled neutronics-thermal-hydraulics PEBBED-THERMIX code. The model uses 71 spectral zones, 35 of which are in the core. In one instance there are no control rods (none are specified in the widely disseminated benchmark based on the PBMR-400 concept). A control rod curtain is added in the second model. The needed converged diffusion theory data (cross sections and diffusion coefficients) are obtained after 3 or 4 iterations between PEBBED-THERMIX and the COMBINE codes. The different numbers of iterations correspond to the different spectral zones. The results for the PBMR-400 model with and without control rods, summarized in Table 1, are discussed below.

In the case without control rods, the model described above yielded an effective multiplication factor of 1.035. Although this is significantly higher than unity, the value is plausible because the average discharged burnup reached in the model corresponds to six passes for the pebbles through the core. The number of passes and the average discharge burnup are not adjusted to give an exactly critical core. In doing so, this study conforms to the six passes through the core for the pebbles, as assumed in the PBMR-400 benchmark currently under development in an OECD/NEA-sponsored international effort. In a design calculation or for an operating reactor, the actual discharge burnup and the number of recycling times of the pebbles through the core are iteratively adjusted to give a multiplication factor of exactly one. The difference in discharge isotopics that result from this difference between the model and the operating reactor is inconsequential for this study, as explained later.

Table 1: Fully Burnt PBMR-400 Spent Fuel Discharge Isotopics

PEBBED-THERMIX-COMBINE Fully Burnt PBMR-400 Spent Fuel Discharge Isotopics					
	Without Control Rod Curtain		**With Control Rod Curtain**		**Confidence Range for Control Rod Curtain Case**
k_{eff}	1.035		1.03583		1.0
	Isotopics (mg/pebble)		**Isotopics (mg/pebble)**		**Discharge Range**
Isotope	**Discharge**	**Fresh**	**Discharge**	**Fresh**	**(mg/pebble)**
U-235	195.6	863.9	198	863.9	185 ± 11
U-238	7639	8134	7645	8134	7690 ± 50
Pu-238	2.6		2.5		2.8 ± 0.2
Pu-239	65.6		66		57 ± 9
Pu-240	33.2		33		30 ± 3
Pu-241	31.1		31		28 ± 4
Pu-242	21.9		21		20 ± 2
Total Pu	154.4		153.5		140 ± 20

In the case of the model with a control rods curtain, the effective multiplication factor is 1.03583. This is larger than for the case without control rods. This counterintuitive result is easily explained by the same argument given above about the cycling of pebbles through the core and the average burnup that is achieved. When control rods are present, the burnup achieved is lower than in the absence of control rods. Since the burnup cutoff and the number of pebble recycling times through the core are not adjusted to produce an exactly critical reactor, the presence of the control rods merely shifts the reactivity to another zone within the reactor. Needless to say, if one starts with an exactly critical reactor with a specified burnup cutoff for the pebble and a corresponding control rod insertion pattern (or with no control rods in), the introduction of control rods (other than their further insertion) would result in a reactivity decrease. In order to operate with control rods in (or with a different control rod insertion pattern), a search must be performed on the recirculation pattern of the pebbles and the burnup cutoff must be adjusted. It is clear from these considerations that the burnup discharge and the isotopics shown in Table 1 are for non-critical configurations of the model reactor and that consequently the computation of the isotopics was performed in a non-critical spectrum. This is expected to result in some discrepancy from the actual reactor operating in a critical reactivity state. This departure is unimportant for this study, however, as shown in the last column of Table 1. That column contains the range of discrepancy between our model results and those obtained with an exactly critical model supplied to us by PBMR [viii]. The actual PBMR information is proprietary but the ranges given in the table are fully representative without disclosing the PBMR data. Although the range of discrepancy shown between the non-critical and the critical spectrum data seem significant, for the purpose of answering the question about the termination of safeguards on the spent fuel they are actually small. This is because even at the limits of the range the criteria for continuation of safeguards are not challenged, as shown later.

The isotopics for the two cases considered here, and shown in the table, are reasonably similar. For all cases the Pu-238 content is very far from the 80% content required for termination of safeguards on measured discards. The fissile nuclides contents per pebble are small but still sufficiently large to be attractive to a would-be proliferant.

The plutonium content of the average spent fuel pebble is large enough to be of interest to a would-be proliferant. Using the information in Table 1, the Pu-239 content per pebble is found to range from as high as 66 mg to as low as 48 mg. The total plutonium content ranges from 120 mg to 160 mg per pebble. Therefore, approximately between 151,516 and 208,333 pebbles are needed to accumulate 10 kg of Pu-239 (ignoring the others isotopes that are present) and between 62,500 and 83,333 pebbles to accumulate 10 kg of plutonium regardless of isotopic distribution. Since the reactor requires approximately between 480 and 500 fresh pebbles per day and discharges that many as fully spent, the accumulation of 10 kg of total Pu-239 in the spent fuel of ***one*** PBMR-400 reactor will require between 303 days and 434 days. It would take between 125 and 174 days to accumulate 10 kg of total Pu, without regard to isotopic distribution.

3.3 Comparison to LWR Spent Fuel

The Pu-238 fraction of total plutonium is approximately 1.7% for the no-control rod case and about 1.6% for the case with control rods in. The value could be as high as 2.5% when criticality is enforced during depletion and isotopics determination. These results show that the Pu-238 fraction of total Pu in PBMR-400 spent fuel is of the same order of magnitude as that expected in typical light water reactor spent fuel. Indeed, in light water reactors spent fuel the Pu-238 content can range from about 1% of the total Pu (see Fig 11 in Ref. [ix]) to as high as 2% [ix, x], depending on the length of the cycle.

3.4 Comparison of PBMR-400 Discharged Fissile Contents to FRESH LWR Fuel Fissile Content

The fissile contents of the PBMR-400 spent fuel are comparable to those of fresh light water reactor fuel. For the case without control rods, the fissile isotopes content is about 4.38 wt-percent if one considers all plutonium isotopes (since they are all fissile) or about 3.27 wt-percent if only Pu-239 and U-235 are considered. Since these contents include Pu-239 in addition to the residual U-235, they correspond to nominally higher enrichment levels (i.e., the 4.38 wt-percent fissile content corresponds to more than 4.38 wt-percent of U-235 enrichment in fuel that uses solely uranium as its fissile material). In computing these values, the ratio of mass of fissile materials to the mass of all the Pu and U isotopes was formed. For the model with the control rod curtain, the fissile contents are 4.4 and 3.3 wt-percent respectively, depending upon whether all isotopes of Pu are included in the fissile inventory or if only U-235 and Pu-239 are accounted for. The corresponding results obtained with a critical spectrum are 3.6 wt-percent and 2.8 wt-percent, respectively.

All of these fissile materials contents predicted for the spent fuel have roughly the same magnitude as the fissile content of fresh light water reactor fuel. As a consequence, with a small amount of processing the PBMR-400 spent fuel could be used as source of fuel for another reactor, albeit not another PBMR.

For completeness, it must be noted that a preliminary assessment shows that the spent fuel from a prismatic block HTR is expected to contain between 5 and 6 ^{wt}/o of fissile material (approximately 4^{wt}/o of U-235 and slightly over 1^{wt}/o of Pu=239) [xi]. Such content is fully suitable for use as a fuel in a LWR, provided the fission products are separated.

3.5 Conclusion on Isotopic Contents

The Pu-238 content of the plutonium with the PBMR-400 spent fuel is of the order of a few percent at most (even when considering isotopics obtained with a truly critical spectrum). This is far below the threshold of 80% specified in applicable IAEA documents for the termination of safeguards. It follows that the PBMR-400 spent fuel should be maintained under safeguards until it is altered appropriately. Furthermore, the fissile isotopes content of the PBMR-400 spent fuel is comparable to the fissile content of ***fresh*** light water reactor fuel. It follows that the PBMR-400 spent fuel could be used as the fresh fuel of another reactor provided some intermediate processing is carried out. That processing would involve the removal of the fission products and could involve the removal of the minor actinides if a future use in a thermal spectrum is considered. The isotopic contents of PBMR spent fuel are such that the fuel could still be recycled and used in the production of power, and also of neutrons, in a nuclear reactor.

4. Reprocessing: Pu Recovery from TRISO Fuel

Fuel Cycle Work has been a main area of focus at the INL for many decades. Various teams at the INL have experience developing and applying reprocessing technologies for every type of commercial nuclear fuel as well as for other less common fuels, such as experimental fuels and fuels used by the US Navy. This experience is directly relevant to the understanding and assessment of the ease (or difficulty) of extraction of plutonium from PBMR spent fuel.

4.1 Status of PBMR/TRISO fuel reprocessing technology

Until as recently as 2002 [xii], it was believed and publicly stated that "there is neither a developed process nor capability anywhere in the world for separating the residual fissionable material from GT-MHR spent fuel." Since the GT-MHR spent fuel is constituted of TRISO particles in a graphite matrix, the same conclusion should equally be valid, that no technology exists for reprocessing the PBMR-400 TRISO-based spent fuel. More recently, it has been increasingly believed that the reprocessing of TRISO-based spent fuel is possible, though difficult and potentially very expensive. Even General Atomics now acknowledges the feasibility of TRISO-based spent fuel reprocessing [xiii]. Others are more definite about the feasibility of reprocessing such spent fuel.

The principal difficulty associated with the reprocessing of TRISO-based spent fuel stems from the nature of its constituents. The fuel proper within the TRISO particle (i.e., UO_2 in the current design) poses no problem beyond those present in light water reactor spent fuel. The difficulties arise because of the chemically inert nature of the graphite and silicon carbide constituents. Because of their inertness, few chemical agents are capable of dissolving them efficiently and in large quantities [xiv]. The known physical and chemical processes for achieving the goal of dissolving the TRISO-based spent fuel were reviewed by Greneche [xiv]. A combination of physical and chemical processes can, at great cost, liberate the desired fuel from its surrounding materials. The processes are possible but expensive, and still under research and development.

The findings discussed above are corroborated by internal INL knowledge. Indeed, at the INL, it has been known for some time, but not externally publicized, that the bench scale and prototype scale reprocessing of TRISO-based spent fuel is possible and that a move to industrial scale reprocessing could be achieved, albeit the process still remains an expensive one to implement.

In all cases, the reprocessing of TRISO-based spent fuel is a difficult and expensive undertaking. Notwithstanding the expense, the fissile content of TRISO-based spent fuel cannot be viewed as irrevocably irretrievable from the point of view of safeguards.

4.2 Prospects and Speculations about Future of TRISO Fuel Reprocessing

The chemical processes involved in the reprocessing of TRISO-based spent fuel are conceptually the same as those needed for reprocessing LWR spent fuel. The difference between the two types of fuel pertains to the breaching of the cladding versus removing the graphite matrix and cracking of the SiC layer in the TRISO particle. The publicly known methods for achieving these latter two tasks were reviewed by Greneche [xiv] and referred to above. Improvements for the existing processes are being actively researched. Yet other processes are possible (as stated above for the INL). In addition, recent community rumors seem to indicate that a new process based on electric arc cracking of the SiC layer may result in an increase in throughput and a significant cost reduction for the overall process. It is therefore legitimate to recognize that it is only a matter of time before an efficient and cost effective process will become known and available for implementation.

4.3 Conclusions on Plutonium Recovery Technologies

The recovery of the plutonium could be expensive but not necessarily prohibitive. The plutonium contained in the PBMR-400 spent fuel cannot be considered to be irretrievable. Furthermore, the Pu-239 content of the spent fuel, though small, is sufficient to be of interest to a would-be proliferant, especially in the case of an outright diversion of large numbers of spent fuel pebbles.

5. Scenarios not Covered and Method Limitations

5.1 Scenarios not covered

This study centered on assessing the strategic value of spent fuel from the PBMR-400 reactor. Here spent fuel meant *fully* burnt fuel that is stored in the spent fuel storage facility. However, in the reactor plant design, a non-fully burnt fuel pebble can also be removed from the reactor and eliminated from further cycling if it has lost its physical integrity. For example, broken pebbles, or strongly abraded pebbles would be automatically removed from the flow stream of pebbles by the "singulizer" after the core discharge chute. Such irradiated fuel pebbles would likely have a greater strategic value than the fully burnt spent fuel, but were not studied in this project. Such irradiated fuel with intermediate burnup should be considered in any safeguard arrangements.

This study considered only the "average" pebble, which is assumed to have completed six passes through the reactor in paths that are representative of the average neutronic behavior of the core. The study did not consider the possibility of fewer than six passes or the passage through selective paths. The study also ignored the possibility of the presence of target pebbles intended for production of plutonium or other material of potential use by a would-be proliferant. Other areas of concern not addressed here include the possibility of materials irradiation outside the core region and the dual use of the core with target fertile pebbles and/or neutronic boosters that do not end up in the spent fuel bins.

5.2 Limitations of Methods

The present study used the current version of the PEBBED code. Therefore, it was limited to assuming that the PBMR-400 reactor under consideration has reached the asymptotic mode. This implies the burnup and fuel distributions are also the asymptotic ones. The approach to asymptotic period may be characterized by slightly different spectra and, therefore, by slightly different discharge isotopics. However, within the confidence limits indicated in the results section, the conclusion of this study remains valid regardless of the status of the reactor away from asymptotic mode. The PEBBED methodology is currently being modified to treat the approach to asymptotic, but that is beyond the scope of this project. Another limitation in the study is the explicit modeling of only the relevant isotopes within the PEBBED code. That is to say, not all nuclides are tracked. Finally, it must be noted that the cross section preparation methodology is still being refined in order to achieve full fidelity. At present, the cross section methodology conforms to the state of the art with its limitations.

6. Conclusions and Recommendation

The principal conclusion of this study is that the fully burnt spent fuel from the PBMR-400 reactor does not meet the IAEA criteria for termination of safeguards on measured discards. Therefore safeguards should be maintained on such spent fuel until it has been treated to meet said IAEA criteria. For completeness, it is important to note that spent fuel from prismatic block HTRs would have to be subjected to at least the same restrictions.

Plutonium in significant quantities is present in the fully burnt spent fuel from the PBMR-400 reactor. Although the isotopic distribution in the spent fuel is not ideal for the manufacturing of nuclear explosives, it is comparable to the distribution in the spent fuel of LWRs, from which it has been argued that nuclear explosives could be manufactured [xv]. Furthermore, the plutonium in the PBMR-400 spent fuel, with a Pu-238 content of less than 2% of total Pu, does not contain anywhere near the

80% of Pu-238 required for termination of safeguards. The estimates obtained in this study indicate that enough plutonium for one bare critical mass could be accumulated in less than a year and possibly as fast as 125 days. The extraction of the materials would require a formidable reprocessing effort with expensive (and visible) environmental controls. Reprocessing the spent fuel from the PBMR-400 is currently possible but extremely difficult and has never been demonstrated at the production scale. However, ongoing research and development are very likely to remove, or at least lower, this difficulty.

The spent fuel from the PBMR-400 could be useful to a nuclear explosives procurement effort in ways other that simply separating its contained plutonium. Indeed, the fissile materials content of the spent fuel is comparable to that of *fresh* fuel for a light water reactor. Therefore, fuel for another reactor could be manufactured starting from fully burnt PBMR-400 spent fuel, albeit with the implementation of necessary chemical separation steps to remove fission products. Even the lowest estimate of remaining fissile materials is high enough to be a possible fresh fuel source for a new rector, especially if the use of a heavy water or beryllium reflector is an option.

From this study, it is abundantly clear that safeguards on the PBMR-400 spent fuel should be maintained until the spent fuel has been treated to remove the fissile materials from it and the residual wastes satisfy the IAEA criteria for the termination of safeguards on measured discards. The residual materials could never satisfy the 80% or greater Pu-238 content, but they could eventually satisfy the dilution or irretrievability criterion. Two possible ways to achieve this end could be pursued: chemical treatment to remove the fissile materials and/or burning the fissile materials in an auxiliary reactor.

7. References

i. Fuks, W. F., C. Viljoen, C. Stoker, C. Koch and E. H. Mathews, “The Interim Fuel Storage of the PBMR Facility,” 2nd International Topical Meeting on High Temperature Reactor Technology, Beijing, CHINA, September 22-24, 2004, #Paper D13.

ii. N. Khlebnikov (IAEA), Letter to Mike Farnitano (ISPO Liaison, Vienna), 2004-08-30.

iii. Terry, W. K., H. D. Gougar, and A. M. Ougouag, "Direct Deterministic Method for Neutronics Analysis and Computation of Asymptotic Burnup Distribution in a Recirculating Pebble-Bed Reactor," Annals of Nuclear Energy 29 (2002) 1345 –1364.

iv. Gougar, H. D., A. M. Ougouag, Richard M. Moore, and W. K. Terry, “Conceptual Design of a Very High Temperature Pebble-Bed Reactor,” in Proceedings of Global 2003 (on CD-ROM), embedded topical meeting in the ANS/ENS International Winter Meeting and Nuclear Technology Expo, New Orleans, November 16-20, 2003.

v. Ougouag, A. M., H. D. Gougar, W. K. Terry, R. Mphahlele, and K. N. Ivanov “Optimal Moderation in the Pebble-Bed Reactor for Enhanced Passive Safety and Improved Fuel Utilization” in PHYSOR-2004 - The Physics of Fuel Cycle and Advanced Nuclear Systems: Global Developments, Chicago, Illinois, April 25-29, 2004, on CD-ROM, American Nuclear Society, Lagrange Park, IL. (2004).

vi. Ougouag, A. M., and H. D. Gougar, “Preliminary Assessment of the Ease of Detection of Attempts at Dual Use of a Pebble-Bed Reactor,” Transactions of the Winter 2001 Annual Meeting of ANS, Reno, NV, Trans. ANS 85, pp. 115-117, Nov. 2001. See also Ougouag, A. M., S. M. Modro, W. K. Terry, and H. D. Gougar “Rational Basis for a Systematic Identification of Critical Components and Safeguard Measures for a Pebble-Bed Reactor” Transactions of the Winter 2002 Annual Meeting of ANS, Washington, DC, Trans. ANS 87, pp. 367-368, Nov. 2002.

vii. Ougouag, A. M., W. K. Terry and H. D. Gougar, “Examination of the Potential for Diversion or Clandestine Dual Use of a Pebble-Bed Reactor to Produce Plutonium,” Proceedings of HTR 2002, 1st International Topical Meeting on High temperature Reactor Technology (HTR), Petten, Netherlands, April 22-24, 2002.

viii. Frederik Reitsma (PBMR Pty Ltd.) private communication to H. D. Gougar on October 25, 2005

ix. A. DeVolpi, “Denaturing Fissile Materials,” Progress in Nuclear Energy, Vol.10, No. 2, pp. 161-220, 1982.

x. “Report to the American Physical Society by the Study Group on Nuclear Fuel Cycles and Waste Management,” Review of Modern Physics, Vol. 50, No. 1, Part II, January 1978.

xi. X. Raepset, CEA, Saclay, France, personal communication to A .M. Ougouag, November 24, 2005

xii. Malcolm P. LaBar, “The Gas Turbine-Modular Helium Reactor: A Promising Option for Near Term Deployment,” GA-A23952, General Atomics Project 04962, April 2002 (also published as a paper in International Congress on Advanced Nuclear Power Plants, and embedded topical meeting at the American Nuclear Society 2002 Annual Meeting, June 9-13, 2002, Hollywood, Florida)

xiii. F. Venneri and F. M. Campbell, “Sustainable Long Term Nuclear Power; The Large Scale deployment of MHRs,” presentation handout, General Atomics, November 2005; personal communication from F. Venneri to A. M. Ougouag, Dec. 7, 2005.

xiv. D. Greneche and P. Brossard, “The Reprocessing Issue for HTR Spent fuels,” Proceedings of ICAPP ’04 Pittsburgh, PA USA, June 13-17, 2004, Paper 4149, pp. 435-444.

xv. J. Carson Mark, “Explosive Properties of Reactor-Grade Plutonium,” *Science and Global Security*, 1992, Vol. 3, pp. 1-13, © 1992, Gordon and Breach Science Publishers S. A.